

Quantitative Infrared Spectroscopy of Minor Constituents of Earth's Atmosphere

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Infrared spectroscopic techniques have become extremely powerful tools for use in achieving a number of observational objectives in understanding and monitoring the "health" of Earth's atmosphere. Prerequisite to designing appropriate instruments as well as to interpreting the observations that monitor the important molecular species, quantitative laboratory spectroscopic measurements must be done. The measurements provide (1) line and band intensity values that are needed to establish limits of detectability for as yet unobserved species and to quantify the abundance of those species that are observed; (2) line positions, half-widths, and pressure-induced shifts, all of which are needed for remote and in situ sensing techniques; and (3) data on these basic molecular parameters at temperatures and pressures appropriate to the real atmosphere.

Good progress has been made in calibrating the BOMEM Fourier Transform Spectrometer (FTS) intensity determinations with measurements obtained with the Kitt Peak interferometer on carbon dioxide (CO_2). The analysis for line intensities has begun for nitric acid (HNO_3) using infrared spectra previously obtained.

An experimental study was completed which demonstrates that magnetic rotation spectroscopy can detect free radical molecular species (in situ) in the part-per-trillion mixing ratio range. In connection with this work, measurements were done to establish

Zeeman tuning rates in the infrared spectrum of nitrogen dioxide (NO_2).

Preliminary measurements have been made to measure line intensities and shapes of gaseous water in the 1-millimeter spectral region to help in understanding the apparent anomaly associated with absorption of solar radiation by clouds in Earth's atmosphere. The signal-to-noise ratio (S/N) on these spectra, recorded with the high-resolution BOMEM FTS and 25-meter base-path multiple-traversal absorption cell, were greatly enhanced by incorporating an infrared band-limiting filter wheel with S/N ratios greater than 1000 achieved in 30 minutes of integration.

This research was performed in collaboration with Linda Brown, Jet Propulsion Laboratory/California Institute of Technology; Mike Dulick, National Solar Observatory; Guy Guelachvili, University of Paris XI; Sumner Davis, University of California, Berkeley; Aaron Goldman, University of Denver; Ginette Tarrago, University of Paris XI; Nelly Lacome, University of Paris VI; Tom Blake, Battelle, Pacific Northwest Laboratory; and Chris Mahon, Space Physics Research Institute.

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Spreading and Growth of Contrails in a Sheared Environment

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The evolution of persistent contrails (condensation trails) has been modeled over time scales of 15–180 minutes using a large-body simulation model with detailed microphysics. Model results have been compared to satellite and in situ measurements of persistent contrails from the Subsonic Aircraft: Contrail and Cloud Effects Special Study. In particular, the evolution of the persistent contrail observed on May 12, 1996, was simulated. In simulations

with large ambient supersaturations and moderate wind shear, crystals with lengths greater than 200 microns are generated within 35 minutes by depositional growth. In situ measurements in the May 12 contrail case showed that these large crystals did in fact form. The large crystals fall rapidly and the contrail's horizontal extent increases as a result of wind shear (see figure). Strong radiative heating (with

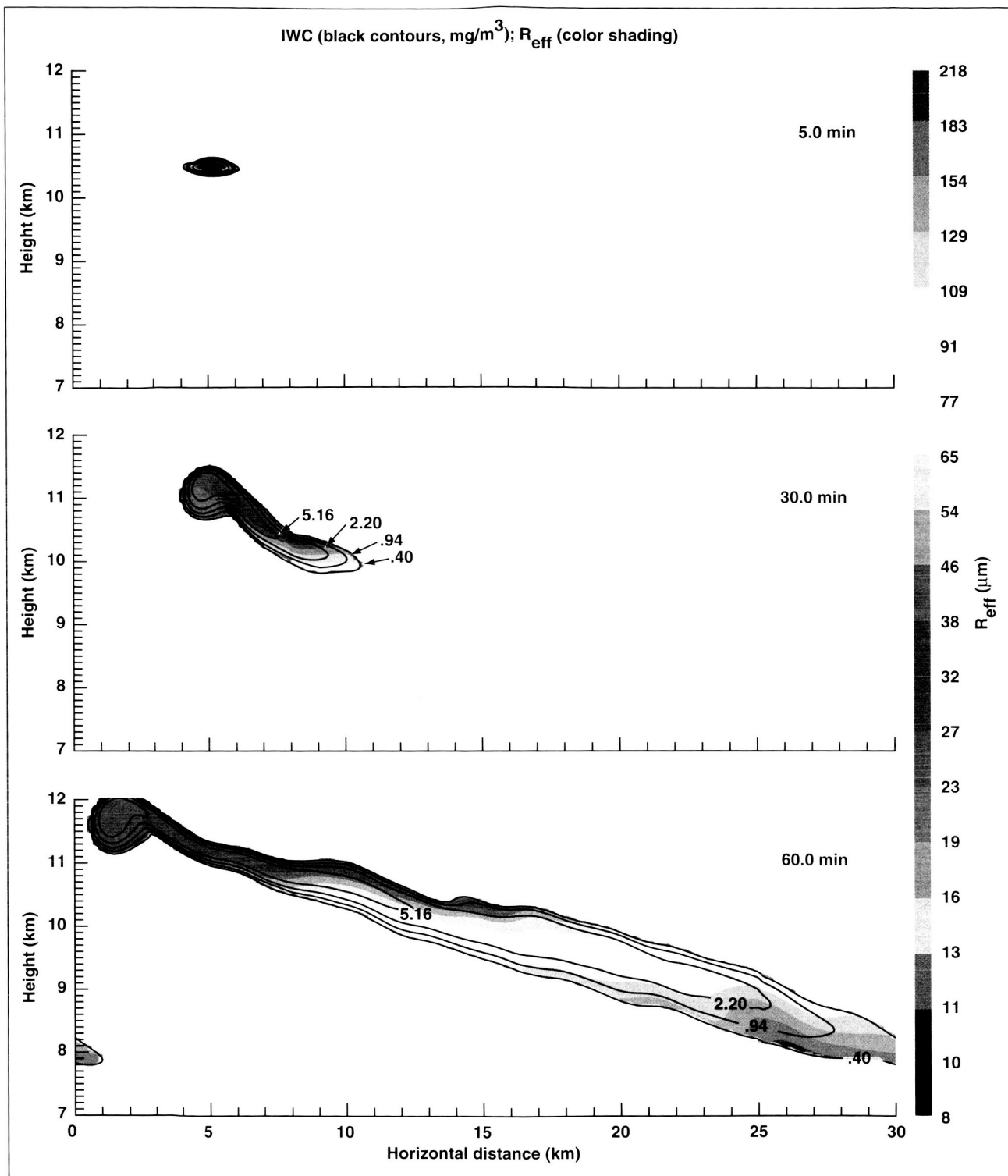


Fig. 1. Contours of ice water content (IWC) are plotted versus cross-track horizontal distance and height at three different times in the baseline simulation. The color shading shows the effective radius (R_{eff}) of the ice crystals. The sorting of crystal size with height as well as the contrail spreading are evident.

rates up to 30 kelvin per day) drives a local updraft and lofts the contrail core several hundred meters. The observed rate of contrail spreading and maintenance of optical depths larger than 0.1 can be explained simply by the growth and precipitation of ice crystals that nucleate during the initial contrail formation if the environmental humidity is high enough (relative humidity with respect to ice greater than 125%). This result is consistent with the high humidities observed in regions where the persistent

contrails formed on May 12. Also, the simulations indicate that the humidity must be high throughout a depth of at least several hundred meters below the contrail to allow the crystals to continue growing as they fall.

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The Roles of Aerosols in Stratospheric Ozone Chemistry

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Stratospheric aerosol can affect the environment in three ways. Sulfuric acid aerosols have been shown to act as sites for the reduction of reactive nitrogen and chlorine and as condensation sites to form (under very cold conditions) polar stratospheric clouds, which facilitate ozone depletion. Recently, modeling studies have suggested a link between black carbon aerosol (BCA) and ozone chemistry. These studies suggest that nitric acid (HNO_3), nitrogen dioxide (NO_2), and ozone (O_3) may be reduced heterogeneously on BCA particles. The ozone reaction converts ozone to oxygen molecules, while HNO_3 and NO_2 react to form nitrogen oxide (NO_x). Finally, a buildup of BCA could reduce the single-scatter albedo of aerosol below a value of 0.98, a critical value that has been postulated to change the effect of stratospheric aerosol from cooling to warming. Correlations between measured BCA amounts and aircraft use have been reported. Attempts to link BCA to ozone chemistry and other stratospheric processes have been hindered by questions concerning the amount of BCA that exists in the stratosphere, the magnitude of reaction probabilities, and the scarcity of BCA measurements.

Recently the Cloud and Microphysics Group participated in the NASA-sponsored Photochemistry of Ozone Loss in the Arctic Region in Summer (POLARIS) mission, which studied ozone depletion mechanisms in the Arctic summer and was completed in Alaska in 1997. The Ames Wire Impactors (AWI) were used in this mission as part of the

complement of experiments on the NASA ER-2 aircraft, with the objective of measuring the character of the stratospheric aerosol during POLARIS and providing this information to the scientific community. A main objective of the study was to determine the amount of aerosol surface area, particularly that of BCA, available for reaction with stratospheric constituents and to assess, if possible, the importance of these reactions. The AWI collect aerosol and BCA particles on thin palladium wires that are exposed to the ambient air in a controlled manner. The samples are returned to the laboratory for subsequent analysis. The product of the AWI analysis is the size, surface area, and volume distributions, and the morphology and elemental composition of the aerosols and BCA. Modifications to the AWI data analysis procedures were required in which the collection of BCA is modeled as a fractal aggregate. The new analysis results in an increase in BCA surface area of approximately 24 and an increase in mass of 10 from the previous method. The character of the BCA and its area distributions measured during POLARIS are compared with those made by the AWI on past missions; the comparisons show trends in the spatial and temporal distribution. One trend confirms that the amount of BCA in the Northern Hemisphere is much greater than in the Southern Hemisphere. For the current study, BCA surface area is used in computer models that attempt to predict measured nitrogen oxide/reactive nitrogen (NO_x/NO_y) ratios.